

# Experimental valley qubit state tomography and coherence induced uncertainty relations in monolayer WSe<sub>2</sub>

Yu-Ran Zhang,<sup>1,2</sup> Jing Wang,<sup>1,2</sup> Chuanrui Zhu,<sup>1,2</sup> Heng Fan,<sup>1,2,3,\*</sup> and Baoli Liu<sup>1,†</sup>

<sup>1</sup>*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

<sup>2</sup>*School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China*

<sup>3</sup>*Collaborative Innovation Center of Quantum Matter, Beijing 100190, China*

Valleytronics as a new electronic conception is an emerging exciting research field with wide potential applications, which is attracting great research interests for their extraordinary properties [1–5]. The localized electronic spins by optical generation of valley polarization [6–9] with spin-like quantum numbers [10–12] are promising candidates for implementing quantum-information processing in solids. It is expected that a single qubit preparation can be realized optically by using combination of left- and right-circularly polarized lights [6–8]. Significantly in a series of experiments, this has already been well achieved by linearly polarized laser representing equal weights of left- and right-circular components resulting in formation of a valley exciton [13–16] as a specific pseudo-spin qubit with equal amplitudes for spin up and spin down. Further researches on the control of valley pseudospin using longitudinal magnetic field [2, 3] and optical Stark effect [1] have been reported. However, a general qubit preparation has not yet been demonstrated. Moreover as a platform for quantum information processing, the precise readout of a qubit state is necessary, for which the state tomography is a standard method in obtaining all information of a qubit state density matrix.

Here, we will lay all necessity foundations in quantum-information processing for the valley pseudospin as a qubit. Our main results are as follows: we show that an arbitrary qubit preparation can be implemented by using the specific elliptical light excitation with combination of the corresponding left- and right-circularly polarized lights. Then we show that the valley qubit readout of the valley pseudospin can be achieved precisely by the standard state tomography by measuring both linear and circular polarization degrees and intensities of the polarization-resolved photoluminescence (PL). Thus the density matrix of the valley qubit is presented quantitatively. To confirm the coherence of the valley qubit, we show that the valley pseudospin qubit can demonstrate the Heisenberg uncertainty principle which the unique characteristic of quantum mechanics, including two different forms: entropic uncertainty relations and Heisenberg uncertainty relations. Our results pave the way for the valley pseudospin as a qubit which is the fundamental element acting as the carrier of quantum information.

**Valley pseudospin as a qubit and its detection by photoluminescence.** Tungsten diselenide (WSe<sub>2</sub>), a two-dimensional ML of TMDs (MX<sub>2</sub>), have emerged as an exciting platform for opto-electronics and quantum information processing, see Fig. 1(a). In particular, a pair of degenerate

energy extremals are present at the  $K$  and  $K'$  valleys in the momentum space of hexagonal MX<sub>2</sub> MLs. These valleys represented by a binary pseudospin behave like a spin-1/2 system where the neutral exciton in the  $K$  valley can be labelled as valley-pseudospin up, and the neutral exciton in the  $K'$  valley can be labelled as valley-pseudospin down [2]. The quantum states of two valleys and their coherent superposition constitute the valley pseudospin, promising the valley degree of freedom to realize a qubit. As optically driven spintronics, the generation and detection of valley pseudospin depend on the facts that the left-circularly ( $\sigma^+$ ) or right-circularly ( $\sigma^-$ ) polarized light selectively excites an electron-hole pair at the  $K$  or  $K'$  valley. In turn, the resulting valley-polarized exciton exclusively couples to left-circularly or right-circularly polarized light and can be detected by polarization-resolved PL, see Fig. 1(b).

In our experiment, the ML WSe<sub>2</sub> flakes are prepared by mechanical exfoliation of bulk WSe<sub>2</sub> crystal (2D semiconductors USA) on SiO<sub>2</sub>/Si substrate. The samples are mounted in a temperature-controlled (4.7 K  $\sim$  300 K) He continuous flow cryostat (Oxford MicrostatHiResII). The measurements are carried out in confocal microscope with a super-long working distance (22 mm) objective (Nikon CFI60-2, 50 $\times$ ). The typical spot size of focused laser is around  $\sim 2 \mu\text{m}$  and is smaller than the size of ML WSe<sub>2</sub>. A 671 nm continuous wave (cw) solid-state laser was used as an excitation light source cleaned by a 671 nm laser line filter. The valley pseudospin polarization is realized by excitonic robust bound states consisting of electrons and holes localized in the  $K$  and  $K'$  valleys, and it can be optically manipulated and detected. The target valley states are initialized by elliptically-polarized excitation laser light passing through a Soleil Babinet Compensator.

The circular basis consists of  $|K\rangle$  and  $|K'\rangle$  valley states which are at two poles in Bloch sphere representation, see Fig 1(c). In the frame of Bloch sphere, an arbitrary pure valley state is written as

$$|\psi_{\theta,\varphi}\rangle = \cos\frac{\theta}{2}|K\rangle + \sin\frac{\theta}{2}e^{i\varphi}|K'\rangle. \quad (1)$$

In order to prepare this state, we need to excite the system by the incident elliptically polarized light superposed by left- and right-circularly polarized lights with different weights and relative phase, in which the weight and phase correspond to  $\theta$  and  $\varphi$ , respectively. The linearly polarized basis consists of two arbitrary orthogonal states located in the equator of Bloch sphere represented by  $\sigma^{X,Y}$  corresponding to valley states  $|\psi_{\frac{\pi}{2},\frac{\pi}{2}\mp\frac{\pi}{2}}\rangle = (|K\rangle \pm |K'\rangle)/\sqrt{2}$ , respectively.

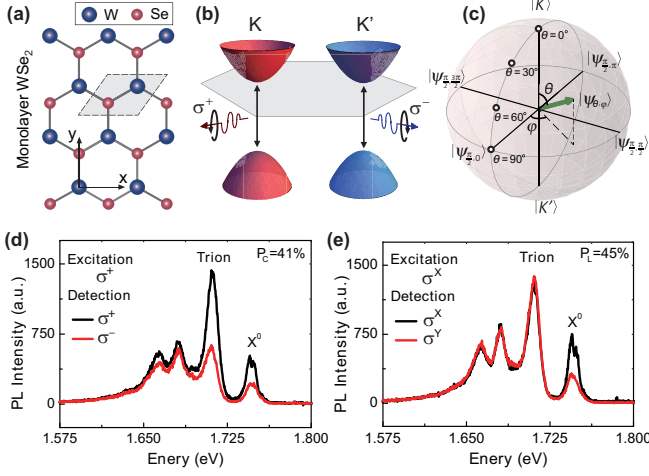


FIG. 1: **Valley-orbit coupled neutral exciton  $X^0$  as a qubit.** (a) The 2D hexagonal crystal structure of a monolayer (ML) transition metal dichalcogenide (TMD) composed of W atoms (blue) and Se atoms (red). (b) Valley optical selection rules for neutral exciton  $X^0$  in ML TMD. In  $K$  and  $K'$  valleys, the neutral excitons  $X^0$  recombine to emit  $\sigma^+$  and  $\sigma^-$  circularly polarized photons, respectively. (c) Bloch sphere representation of the optical manipulation of neutral excitonic pseudo-spin states. (d) Photoluminescence (PL) spectra of ML WSe<sub>2</sub> at 4.7 K under  $\sigma^+$  circularly polarized excitation. (e) PL spectra of ML WSe<sub>2</sub> at 4.7 K under  $\sigma^X$  linearly polarized excitation.

**Normalized neutral exciton measurement and valley state tomography.** For the measurements of prepared valley states in the basis of  $|K\rangle$  and  $|K'\rangle$  vectors, we detect the left- and right-circularly polarized PL intensities of neutral exciton  $X^0$  and obtain the circular polarization  $\eta_C$  by measuring the circularly polarized components of the emitted PL intensity  $I(\sigma^\pm)$  as [2, 6]  $\eta_C = [I(\sigma^+) - I(\sigma^-)]/[I(\sigma^+) + I(\sigma^-)]$ . Similarly, we can detect the valley linear polarization  $\eta_L$  by measuring the linearly polarized components of the emitted PL intensity  $I(\sigma^{X,Y})$  as  $\eta_L = [I(\sigma^X) - I(\sigma^Y)]/[I(\sigma^X) + I(\sigma^Y)]$ . Figures 1(d,e) show the typical PL spectra of ML WSe<sub>2</sub> at 4.7 K under  $\sigma^+$  circular and  $\sigma^X$  linear polarization excitation, respectively. Both spectra show the higher polarization of neutral exciton  $X^0$ . To initialize any pure valley state (S1), degrees of circular polarization and linear polarization of excitation light, are ideally  $\eta_C = \cos \theta$  and  $\eta_L = \sin \theta$ . In this paper, we have prepared four valley states with  $\varphi = 0$  and  $\theta = 0^\circ, 30^\circ, 60^\circ, 90^\circ$ , respectively. The normalized population of polar projective measurement can be obtained by using experimental degrees of both circular and polarization of PL, see Supplementary Information for details.

Preparation and a complete readout of a qubit state by measurement are necessary for quantum information processing. In general, the readout can be done by using state tomography to obtain all information of the state density matrix. Besides the operators in the poles of Bloch sphere as already been performed in literatures for complete state tomography, we need also the measurement with vectors in the equator to detect the

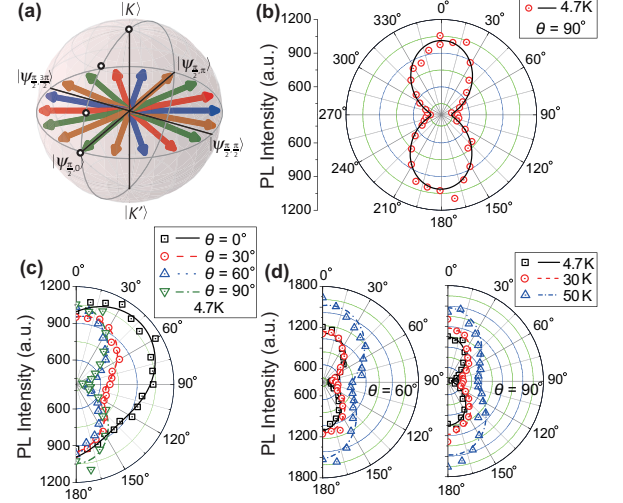


FIG. 2: **Signature of valley quantum coherence.** (a) To detect the excitonic valley quantum coherence, a series of equatorial projective measurements are applied with projectors  $\hat{\Pi}_\alpha \equiv |\psi_{\frac{\pi}{2},\alpha}\rangle\langle\psi_{\frac{\pi}{2},\alpha}|$  and with detection angles  $\alpha$  from  $0^\circ$  to  $360^\circ$ . (b) PL intensity as a function of detection angles  $\alpha$  when the neutral exciton state  $|\psi_{\frac{\pi}{2},0}\rangle$  is excited by  $\sigma^X$  linearly polarized laser at 4.7 K. (c) PL intensity as a function of detection angles  $\alpha$  at 4.7 K for different neutral exciton states  $|\psi_{\theta,0}\rangle$  by linearly polarized laser with  $\theta = 0^\circ, 30^\circ, 60^\circ, 90^\circ$ , respectively. (d) At different temperatures 4.7 K, 30 K and 50 K, PL intensity as a function of detection angles  $\alpha$  where the left diagram is for a neutral exciton state  $|\psi_{\frac{\pi}{2},0}\rangle$  and the right one is for a neutral exciton state  $|\psi_{\frac{\pi}{2},\pi}\rangle$ .

valley quantum coherence and the angle dependent linear polarization PL intensities can be recorded. The states in the equator of the Bloch sphere are generally named as equatorial qubits in quantum information. This measurement corresponds to a series of projectors,  $\hat{\Pi}_\alpha \equiv |\psi_{\frac{\pi}{2},2\alpha}\rangle\langle\psi_{\frac{\pi}{2},2\alpha}|$ ,  $|\psi_{\frac{\pi}{2},2\alpha}\rangle = (|K\rangle + e^{i2\alpha}|K'\rangle)/\sqrt{2}$  with detection angle  $\alpha$  ranging from  $0^\circ$  to  $360^\circ$ , see Fig. 2(a). The z-axis rotation of the valley qubit in the Bloch sphere with an angle  $2\alpha$  will cause the PL polarization to rotate with only the angle  $\alpha$  [1, 2]. Given different valley states, we plot the PL intensity at different temperatures ( $T = 4.7$  K, 30 K, 50 K) as a function of detection angles  $\alpha$  in Fig. 2(b-d). For the projective measurement operator  $\hat{\Pi}_\alpha$  on any pure state  $|\psi_{\theta,\phi}\rangle$ , the measured result could be  $p(\alpha) = [1 + \sin \theta \cos(\varphi - 2\alpha)]/2$ . By normalizing the neutral exciton PL intensity, we can obtain the populations for a series of projective measurements, see Supplementary Information for details.

Then, we can calculate the state tomography results of a valley qubit state. The diagonal elements are obtained by normalized population results of polar projective measurement. The nondiagonal elements are calculated by using the least square method in analyzing the normalized PL results. (See Supplementary Information for details.) With two polar angles  $\theta = 60^\circ, 90^\circ$  and zero azimuthal angle, the state tomography results of valley pseudospin superposition states for different temperatures are shown in Fig. 3.

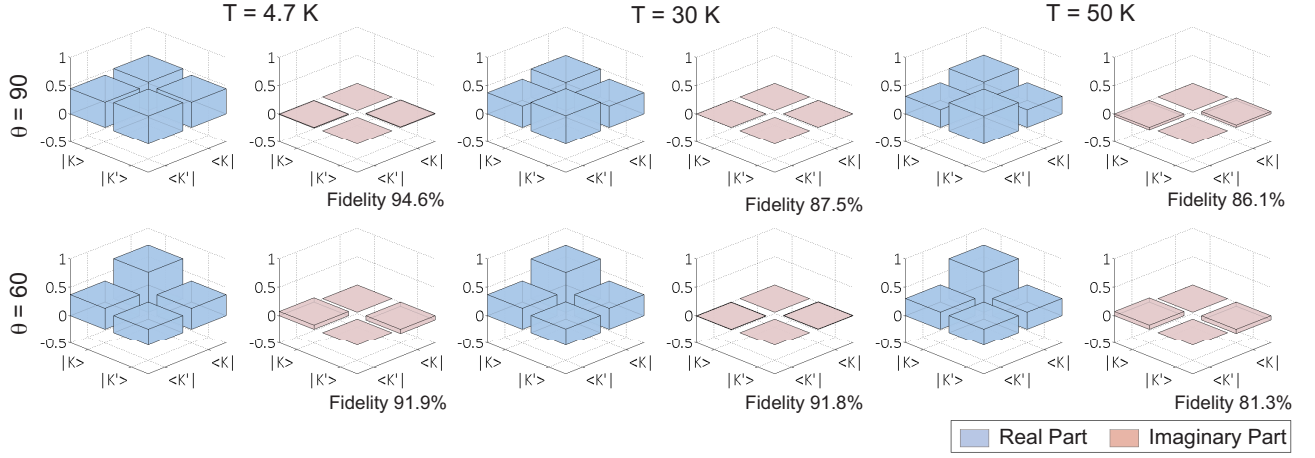


FIG. 3: **Valley state tomography.** The state tomography results of valley pseudospin superposition state for different temperatures  $T = 4.7\text{ K}, 30\text{ K}, 50\text{ K}$  given two polar angles ( $\theta = 60^\circ, 90^\circ$ ) and zero azimuthal angle. The fidelity is defined as  $F(\rho_1, \rho_2) = \text{Tr}(\sqrt{\sqrt{\rho_1}\rho_2\sqrt{\rho_1}})$  for any two states  $\rho_1$  and  $\rho_2$

**Demonstration of uncertainty relations using normalized PL intensity.** One direct verification of the quantum coherence of valley coherence is to demonstrate the uncertainty relations, which are closely related with no-cloning theorem of quantum information [20]. The uncertainty relation that bounds the uncertainties about the outcomes of two incompatible observables on one particle was firstly introduced by Heisenberg using the standard deviation [5], which is the widely accepted form called the Heisenberg-Robertson relation [6]. In an information-theoretic context of quantum information, the uncertainty principle can be formulated as the entropic uncertainty relation [23, 24], which can also be regarded as a quantum coherence [9] induced uncertainty relation, see Supplementary Information for details.

Here, given different valley states, we choose two observables as  $\hat{R} = \hat{\Pi}_0 - \hat{\Pi}_1^\perp$  and  $\hat{Q} = \hat{\Pi}_\alpha - \hat{\Pi}_\alpha^\perp$  with detection angle  $\alpha$  and show the entropy uncertainty  $S(\hat{R}(\rho)) + S(\hat{Q}(\rho))$  (points) against  $\alpha$  compared with the lower bounds and theoretic results (lines) in Fig. 4(a,b), where the forbidden shaded area depends on the lower bound  $\log_2[1/c(\hat{\Pi}_0, \hat{\Pi}_\alpha)]$ . Then, for completeness, we study the Heisenberg uncertainty relation in ML WSe<sub>2</sub>. As  $\hat{R}^2 = \hat{Q}^2 = \mathbb{I}$ , we obtain that  $\Delta_\rho \hat{R}^2 \Delta_\rho \hat{Q}^2 = (1 - \langle \hat{R} \rangle^2)(1 - \langle \hat{Q} \rangle^2)$ , where the average reads  $\langle \hat{Q} \rangle_\rho \equiv \text{Tr}(\hat{Q}\rho) = 2p(\alpha) - 1$  with probability of projector  $p(\alpha) = \text{Tr}(\hat{\Pi}_\alpha \rho)$ , and the derivation is defined as  $\Delta_\rho \hat{R} \equiv (\Delta_\rho \hat{R}^2)^{1/2}$  with variance  $\Delta_\rho \hat{R}^2 \equiv \langle \hat{R}^2 \rangle_\rho - \langle \hat{R} \rangle_\rho^2$ . In Fig. 4(c,d), we plot the Heisenberg uncertainty relation of valley superposition states (points) at different temperatures against the detection angle  $\alpha$  in comparison with the theoretic lower bounds (lines), see Supplementary Information for details.

**Summary and discussion.** In this work, we demonstrate an arbitrary qubit preparation by using the specific elliptical light excitation with combination of the corresponding left- and right-circularly lights. In addition, we achieve the complete readout of the qubit by the state tomography by measur-

ing the circular polarization and intensities of the polarization-resolved PL with respect to different exciton angles. Thus, with this standard tomography technique, the density matrix of the valley qubit can be measured quantitatively. To confirm the quantum coherence of the valley qubit, the unique characteristic of quantum mechanics, we show that the valley qubit can demonstrate the Heisenberg uncertainty principle in two different forms: the Heisenberg-Robertson relation and

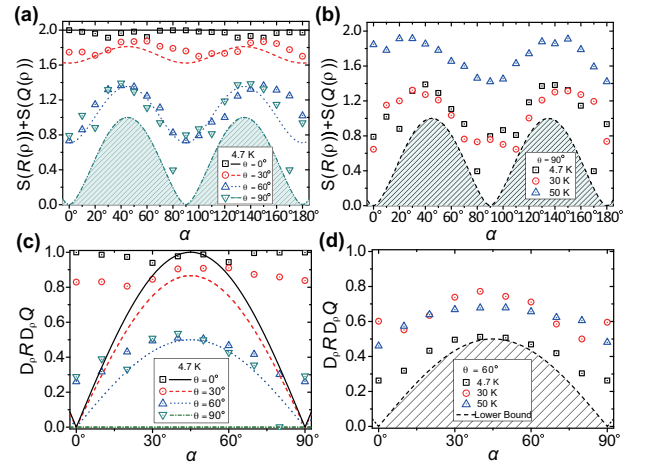


FIG. 4: **Valley uncertainty relations.** (a) Entropy uncertainty  $S(\hat{R}(\rho)) + S(\hat{Q}(\rho))$  against the detection angle  $\alpha$  at  $T = 4.7\text{ K}$  given four valley states,  $\rho = |\psi_{\theta,0}\rangle\langle\psi_{\theta,0}|$  with  $\theta = 0^\circ, 30^\circ, 60^\circ, 90^\circ$ . (b) Given a valley state  $\rho = |\psi_{\pi/2,0}\rangle\langle\psi_{\pi/2,0}|$ , entropy uncertainty  $S(\hat{R}(\rho)) + S(\hat{Q}(\rho))$  against the detection angle  $\alpha$  at different temperatures  $T = 4.7\text{ K}, 30\text{ K}$  and  $50\text{ K}$ . (c) Uncertainty  $\Delta_\rho \hat{R} \Delta_\rho \hat{Q}$  against the detection angle  $\alpha$  at  $T = 4.7\text{ K}$  given four valley states,  $\rho = |\psi_{\theta,0}\rangle\langle\psi_{\theta,0}|$  with  $\theta = 0^\circ, 30^\circ, 60^\circ, 90^\circ$ . (d) Given a valley state  $\rho = |\psi_{\pi/2,0}\rangle\langle\psi_{\pi/2,0}|$ , uncertainty  $\Delta_\rho \hat{R} \Delta_\rho \hat{Q}$  against the detection angle  $\alpha$  at different temperatures  $T = 4.7\text{ K}, 30\text{ K}$  and  $50\text{ K}$ . The forbidden shaded area depends on the lower bound  $|\langle [\hat{R}, \hat{Q}] \rangle_\rho|/2$ .

the entropic uncertainty relation. Our results may pave the way for the valley pseudospin as a qubit which is the fundamental element acting as the carrier of quantum information. With developments on coherent manipulation of valley pseudospin states [2–4, 27–32], more researches by valley qubit can be expected in studying fundamentals of quantum physics and applications in quantum computation and quantum information processing.

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**Authors contributions.** B.L. and H.F. designed the experiment. B.L. is in charge of the experiment, H.F. is in charge of the theory. J.W., C.Z. and B.L. performed the experiment. Y.-R.Z. and H.F. carried out the theoretical study. Y.-R. Z. wrote the paper with assistance of J.W., C.Z., B.L. and H.F. The first two authors J.W. and Y.-R.Z. contributed equally to this paper. All authors analysed the data and commented on the manuscript.

### Additional information

**Competing financial interests:** The authors declare no competing financial interests.

\* Electronic address: hfan@iphy.ac.cn

† Electronic address: blliu@iphy.ac.cn

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## SUPPLEMENTARY INFORMATION

### Normalized projective measurement on the equator of the Bloch sphere

We denote  $|G\rangle$  to the common ground state for excitons in both  $K$  and  $K'$  valleys. The creation and emission of the valley excitons obey the optical selection rules [1]  $\langle G|\mathcal{P} \cdot \hat{\sigma}^+|K\rangle = \langle G|\mathcal{P} \cdot \hat{\sigma}^-|K'\rangle = \mathcal{D}$  and  $\langle G|\mathcal{P} \cdot \hat{\sigma}^-|K\rangle = \langle G|\mathcal{P} \cdot \hat{\sigma}^+|K'\rangle = 0$  where  $\mathcal{P}$  is the electric dipole operator,  $\hat{\sigma}^\pm = (\hat{\sigma}^X \pm i\hat{\sigma}^Y)/\sqrt{2}$  is the circular polarization direction of the electric field, and  $\mathcal{D}$  is the magnitude of the transition dipole moment for both valleys. For any pure state of valley state

$$|\psi_{\theta,\varphi}\rangle = \cos\frac{\theta}{2}|K\rangle + \sin\frac{\theta}{2}e^{i\varphi}|K'\rangle. \quad (S1)$$

and degenerate excitons in  $K$  and  $K'$  valleys without control, the total fluorescence intensity with linearly polarized vector  $\hat{\sigma}^\alpha = \cos\alpha\hat{\sigma}^X - \sin\alpha\hat{\sigma}^Y$  may be written as

$$I_\theta(\alpha) \propto |\langle G|\mathcal{P} \cdot \hat{\sigma}^\alpha|\psi\rangle|^2 = \frac{|\mathcal{D}|^2}{2}[1 + \sin\theta\cos(\varphi - 2\alpha)] \quad (S2)$$

With two general decay mechanisms, we can obtain the collective fluorescence intensity as

$$I_\theta(\alpha) \propto \int_0^\infty dt |\mathcal{D}|^2 [e^{-t/T_1} + e^{-t/T_2^*} \sin\theta\cos(\varphi - 2\alpha)]/2 \\ \propto [1 + (T_2^*/T_1)\sin\theta\cos(\varphi - 2\alpha)]/2 \quad (S3)$$

where  $T_1$  is the exciton population lifetime,  $T_2^* \equiv 1/(1/T_1 + 1/T_2)$  with  $T_2$  the valley exciton coherence time, and the ratio may be calculated as  $T_2^*/T_1 \simeq 0.2$  at 4.7 K. Technically, we can normalize the PL intensity and obtain the probability distribution for the measurement  $\hat{\Pi}_\alpha \equiv |\psi_{\frac{\pi}{2},\alpha}\rangle\langle\psi_{\frac{\pi}{2},\alpha}|$  on state (S1) as

$$p_\theta(\alpha) \equiv (rI_\theta(\alpha) - I_{\frac{\pi}{2}}^{\min})/(I_{\frac{\pi}{2}}^{\max} - I_{\frac{\pi}{2}}^{\min}) \quad (S4)$$

where  $r \equiv (I_{\frac{\pi}{2}}^{\max} + I_{\frac{\pi}{2}}^{\min})/(I_\theta^{\max} + I_\theta^{\min})$ ,  $I_\theta^{\min}$  and  $I_\theta^{\max}$  denote to the minimal and maximal intensities of PL for different detection angles. Compared with the ideal theoretic prediction

$$p(\alpha) = [1 + \sin\theta\cos(\varphi - 2\alpha)]/2, \quad (S5)$$

the experimental results of the equatorial detection probabilities are shown in Fig. S5 for elliptically polarized exciton angles  $\theta = 0^\circ, 30^\circ, 60^\circ, 90^\circ$  at 4.7 K.

### Valley state tomography by normalized PL intensity and circular polarization degree.

For the elliptically polarized exciton light corresponding to the angle  $\theta$ , the intensity of light detected in the left-circularly and right-circularly polarized vectors may be divided to three parts  $I = I_{\text{th}} + I_{\text{pol}} + I_{\text{PL}}$ , where  $I_{\text{th}} = I_1 \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$  denotes to

the coherency matrix of the unpolarized thermal light,  $I_{\text{pol}} = I_2 \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}$  stands for the polarized thermal light and

$$I_{\text{PL}} = I_3 \begin{pmatrix} 1 + \cos\theta & e^{-\Gamma-i\varphi}\sin\theta \\ e^{-\Gamma+i\varphi}\sin\theta & 1 - \cos\theta \end{pmatrix} \quad (S6)$$

corresponds to PL where the valley qubit state may be a mixed state and the temperature decay with a rate  $\Gamma > 0$  is assumed. The intensity of detected light may be described as  $I(\sigma^\pm) = I_1 + I_2 + (1 \pm \cos\theta)I_3$ . The circular PL polarization may be calculated as  $\eta_C = q_3 \cos\theta$ , where the proportion of three different parts of light is defined as  $q_i \equiv I_i/(I_1 + I_2 + I_3)$  for  $i = 1, 2, 3$ . As  $\eta_C \propto \cos\theta$  and using the linear-regression analysis method, the diagonal elements of the density matrix of the valley state are retrieved using the results of circular polarizations of PL for different exciton angles in TABLE S1.

Given the measurement on the equator of the Bloch sphere  $\hat{\Pi}_\alpha \equiv |\psi_{\frac{\pi}{2},2\alpha}\rangle\langle\psi_{\frac{\pi}{2},2\alpha}|$ , the detected PL may be written as

$$I_\alpha(\theta) = I_1 + I_3[1 + \sin\theta\cos(\varphi - 2\alpha)e^{-\Gamma}] \quad (S7)$$

where we have neglected the polarized part of the thermal light ( $I_2 \simeq 0$ ). As discussed in the previous section, we can normalize the PL detected intensity and obtain the normalized probability. One general form of the the probability of equatorial detection may be written as

$$p = \frac{1}{2} + \frac{1}{2}\sin\theta e^{-\Gamma}[\cos(2\alpha)\cos\varphi + \sin(2\alpha)\sin\varphi] \quad (S8)$$

Then, to retrieve the nondiagonal elements of the density matrix of the valley state, we use the least-square method to calculate the real part  $\sin\theta\cos\varphi e^{-\Gamma}/2$  and imaginary part  $\sin\theta\sin\varphi e^{-\Gamma}/2$  given detection angles  $\alpha = 0^\circ \sim 180^\circ$ .

### Demonstration of valley qubit dynamics

Universal control of valley qubit is essential for its various applications in quantum information processing and quantum

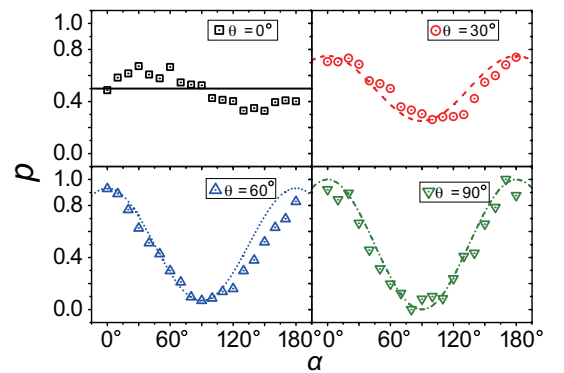


FIG. S5: **Normalized readout of the valley equatorial detection probabilities.** Experimental results of equatorial detection probabilities for elliptically polarized exciton angles  $\theta = 0^\circ, 30^\circ, 60^\circ, 90^\circ$  at 4.7 K, compared with theoretic predictions.



computations. The manipulation of valley pseudospin can be realized using longitudinal magnetic field [2, 3] and optical Stark effect [1]. The valley neutral exciton in ML WSe<sub>2</sub> can be controlled by an external magnetic field that is vertical to the sample plane [2]. For linearly polarized laser excitation  $\theta = \pi/2$  and a longitudinal magnetic field  $B$ , the valley pseudospin evolves with time as  $(e^{-i\Omega t/2}|K\rangle + e^{i\Omega t/2}|K'\rangle)/\sqrt{2}$  and the two valleys become nondegenerate with energy difference  $\Omega = g\mu_B B\hbar^{-1}$  with  $\mu_B$  the Bohr magneton and  $g \simeq -3.7$  the Landé  $g$  factor [4]. Then, the polarization-resolved PL intensity can be written as

$$I(\alpha) \propto \int_0^\infty dt [e^{-t/T_1} + e^{-t/T_2^*} \cos(\Omega t - 2\alpha)]/2 \\ \propto \frac{1}{2} + \frac{T_2^*}{2T_1[1 + (\Omega T_2^*)^2]^{1/2}} \cos(\tilde{\varphi} - 2\alpha). \quad (\text{S9})$$

Thus, we can obtain that the PL intensity under a longitudinal magnetic field is rotated by an angle  $\tilde{\varphi}/2$  with  $\tilde{\varphi} \equiv \arctan(\Omega T_2^*)$  compared with the stationary PL intensity, see Fig. S6(a), from which the details of valley pseudospin dynamics can be retrieved, see Fig. S6(b).

#### Quantum coherence and uncertainty relations.

The uncertainty relation that bounds the uncertainties about the outcomes of two incompatible observables on one particle was firstly introduced by Heisenberg using the standard deviation [5]. One widely accepted form of this relation is expressed by the Heisenberg-Robertson relation [6]:  $\Delta\hat{R}\Delta\hat{Q} \geq |\langle[\hat{R}, \hat{Q}]\rangle|/2$  where  $\Delta\hat{R}$  is the standard deviation of an observable  $\hat{R}$ . As this form of relations is state-dependent on the right-hand-side, an improvement of uncertainty relations, in an information-theoretic context, was subsequently proposed and expressed as [7, 8]  $H(\hat{R}) + H(\hat{Q}) \geq \log_2[1/c(\hat{R}, \hat{Q})]$  where  $H(\hat{R})$  denote the Shannon entropy of the probability distribution of the outcomes when  $\hat{R}$  is measured and

$c(\hat{R}, \hat{Q}) \equiv \max_{j,k} |\langle r_j | q_k \rangle|^2$  given  $|r_j\rangle$  and  $|q_k\rangle$  the eigenvectors of  $\hat{R}$  and  $\hat{Q}$ , respectively.

The relative entropy of coherence [9] takes the form,  $C(\hat{M}) \equiv S(\rho_{\text{diag}}) - S(\rho)$ , where  $\rho$  is the density matrix,  $\rho_{\text{diag}}$  keeps only the diagonal elements of  $\rho$  in a fixed basis  $\hat{M}$ , for example the computational basis  $\{|j\rangle\}$ ,  $S(\cdot)$  is the von Neumann entropy. The physical implication of this definition is that the coherence of a quantum state can be interpreted as the entropy creating in the measurement  $\hat{M}$  corresponding to the fixed basis subtracting the original entropy [9]

$$C(\hat{M}) \equiv S(\hat{M}(\rho)) - S(\rho) \quad (\text{S10})$$

where  $\hat{M}(\rho) = \sum_j |j\rangle\langle j|\rho|j\rangle\langle j|$  is the post-measurement state which is the diagonal matrix in a fixed basis and equals to the Shannon entropy  $H(\hat{M})$ . For projective measurement operator in the equator of Bloch sphere  $\hat{M} = \hat{\Pi}_\alpha$  and initial state (S1), the von Neumann entropy of  $\hat{M}(\rho)$  can be written as a binary entropy  $S(\hat{M}(\rho)) = H_b(p) = -p \log p - (1-p) \log(1-p)$  where  $p = [1 + \sin \theta \cos(\varphi - \alpha)]/2$ . By applying this observation to a set of measurement operators  $\{\hat{M}_k\}$ , we may find that total coherence in different measurements bases is larger than a bound,  $\sum_k C(\hat{M}_k) \geq B(\{\hat{M}_k\}, \rho)$ . For example, using the entropy uncertainty, we can obtain a lower bound of coherence in different basis as

$$C(\hat{R}) + C(\hat{Q}) \geq \log_2[1/c(\hat{R}, \hat{Q})] + 2S(\rho) \geq \log_2[1/c(\hat{R}, \hat{Q})] \quad (\text{S11})$$

where the first equal condition is the same as the one of entropic uncertainty and the second equality is saturated for pure states.

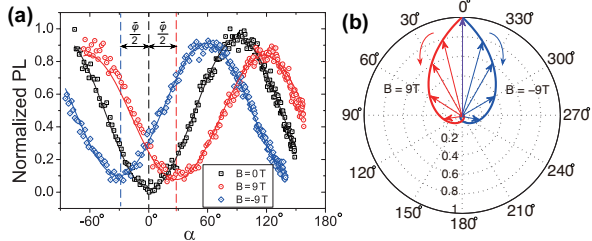


FIG. S6: **Dynamics of valley qubit by a longitudinal magnetic field.** (a) The normalized angle dependent PL intensity of the valley exciton for  $B = 0$  T (black square),  $B = 9$  T (red circle) and  $B = -9$  T (blue triangle). (b) Representation of valley qubit dynamics on the equatorial plane of Bloch sphere for  $B = 9$  T (red) and  $B = -9$  T (blue). The valley qubit state vector is rotated with angular velocity  $\Omega \simeq 2.93 \times 10^{12}$  rad/s. The decoherence rate can be written as  $e^{-t/T_2^*}$  with the effective coherence time  $T_2^* \simeq 0.37$  ps [2].

\* Electronic address: hfan@iphy.ac.cn

† Electronic address: blliu@iphy.ac.cn

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		Polarization $\theta = 0^\circ$	$\theta = 30^\circ$	$\theta = 60^\circ$	$\theta = 90^\circ$
Excitation laser	$\eta_L$	14.3%	50%	86.6%	99.6%
	$\eta_C$	99%	86.6%	50%	8.7%
PL	$\eta_L$	14.3%	20%	36%	41.8%
	$\eta_C$	37.3%	35.4%	16.3%	0%

TABLE S1: Experimental results of degrees of both circular polarization and linear polarization of linear exciton laser and PL.

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